

SEMI-ANNUAL STATUS REPORT  
for a  
THEORETICAL INVESTIGATION OF ISOTOPIC ANOMALY OF  
XENON IN CARBONACEOUS CHONDRITES AND OTHER TERRESTRIAL  
AND EXTRA-TERRESTRIAL SAMPLES.  
NGR 19-011-012 (Supplement No. 2)

by

D. D. SABU  
DEPARTMENT OF CHEMISTRY  
GRAMBLING COLLEGE  
GRAMBLING, LA 71245

March 15, 1974

NASA-CR-138139) THEORETICAL INVESTIGATION  
OF ISOTOPIC ANOMALY OF XENON IN  
CARBONACEOUS CHONDRITES AND OTHER  
TERRESTRIAL AND (Grambling Coll., La.)  
25 p HC \$4.75

CSCL 03B

G3/30 - 16935

N74-22439

Unclas

# SEMI-ANNUAL STATUS REPORT

SUBMITTED TO: NASA Scientific & Technical Information  
Facility  
P. O. Box 33  
College Park, Maryland 20740

REFERENCE NUMBER: NGR-19-011-012 (Supplement No. 2)

INSTITUTION: Grambling College  
Grambling, LA 71245

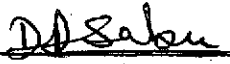
TITLE: Theoretical Investigation of Isotopic  
Anomaly of Xenon in Carbonaceous  
Chondrites and Other Terrestrial and  
Extra-terrestrial Samples.

PRINCIPAL INVESTIGATOR: D. D. Sabu  
Associate Professor  
Department of Chemistry

DURATION: Two (2) years

SUM GRANTED BY NASA: \$21,539

DATE: September 1, 1973

SIGNATURE:   
D. D. Sabu  
Department of Chemistry  
Grambling College  
Grambling, LA 71245

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The present report represents a summary of the work performed subsequent to last semi-annual status report submitted to NASA Scientific and Technical Information Facility on August 10, 1973. The results of our studies are contained in two appendices. Following is a brief summary of the articles contained in the appendix I and II.

Trapped Xenon in Meteorites:

This work done in collaboration with Professor O. K. Manuel and Mr. E. W. Hennecks of University of Missouri - Rolla, has been submitted to Nature. In the previous work (O. K. Manuel, E. W. Hennecke and D. D. Sabu, Nature Physical Science, 240, p. 99-101, 1972) we had pointed out that the xenon released from carbonaceous chondrites between 600° - 1000° C consists of two isotopically distinct components of trapped xenon. This important work (see e. g. Nature, 240, p. 378 - 379, 1972) has been further elaborated in the present paper. We note that the three reservoirs of xenon- solar, atmospheric and meteoritic are not related by simple mass dependent fractionation. Further we note that total xenon in carbonaceous meteorites whose average is represented by AVCC xenon is in fact a varying mixture

of component X and trapped meteoritic xenon which is distinct from AVCC xenon.

The isotopic composition of trapped meteoritic xenon is as follows:

$\text{Xe}^{124} : \text{Xe}^{126} : \text{Xe}^{128} : \text{Xe}^{130} : \text{Xe}^{131} : \text{Xe}^{132} : \text{Xe}^{134} : \text{Xe}^{136} = 0.0276:0.0248$   
 $0.501: 1.00: 5.04: 6.19: 2.31: 1.90.$

Meteoritic trapped xenon is related to solar xenon probably through same set of processes which relate atmospheric xenon to solar xenon. We have tried to explore the relationship in the next paper which is presented in preliminary form in the Appendix - II.

#### Xenon Anomaly: The Possible Role of Mass Fractionation

In 1967 Canals at University of Missouri at Rolla performed an experiment on shale which is one of the richest sources of xenon on earth. He heated South African fig tree shale for 95 hours at  $200^{\circ}\text{C}$  then analyzed the xenon released at  $400^{\circ}\text{C}$  on heating for one hour. We note that Shale- $400^{\circ}\text{C}$  xenon is a mass fractionated xenon. However the pattern of fractionation is not linear which is contrary to popular belief. The pattern of fractionation observed between solar and atmospheric xenon. A mechanism in terms of early incorporation of solar wind xenon in the primordial matter is forewarded to explain the puzzle of trapped Ar and Kr isotopes which are identical in composition in earth, moon and meteorites. This mechanixm is also capable to explain the extraordinarily high relative abundance of Kr in the earth's atmosphere.

## APPENDIX - I

(Paper Submitted to Nature, 1973)

## Trapped Xenon in Meteorites

D. D. Sabu  
Department of Chemistry  
Grambling College  
Grambling, Louisiana 71254, U.S.A.

and

E. W. Hennecke and O. K. Manuel  
Nuclear Division  
Department of Chemistry  
University of Missouri-Rolla  
Rolla, Missouri 65401, U.S.A.

### ABSTRACT

Xenon in meteorites can be resolved into a mixture of component X and trapped xenon with the following isotopic composition,  $^{124}\text{Xe} : ^{126}\text{Xe} : ^{128}\text{Xe} : ^{130}\text{Xe} : ^{131}\text{Xe} : ^{132}\text{Xe} : ^{134}\text{Xe} : ^{136}\text{Xe} = 0.0276 : 0.0248 : 0.501 : 1.00 : 5.04 : 6.19 : 2.31 : 1.90$ . This trapped meteoritic xenon is distinct from AVCC xenon, which is shown to represent the average composition of the total xenon in meteorites containing various mixtures of component X and trapped meteoritic xenon.

In 1960 Reynolds<sup>1</sup> reported the discovery of radiogenic  $^{129}\text{Xe}$ , the decay product of extinct  $^{129}\text{I}$ , in the Richardton chondrite. He also noted a general anomaly pattern across the isotopes of xenon in the Richardton meteorite, and later that year he<sup>2</sup> reported a similar isotopic anomaly pattern in the xenon released from the Murray carbonaceous chondrite. Since Murray contained over 40 times more xenon than Richardton, but less cosmogenic noble gas isotopes than Richardton, Reynolds<sup>2</sup> concluded that the general anomaly pattern was characteristic of primordial xenon.

Following Reynolds<sup>1,2</sup> discovery of general isotopic anomalies in meteoritic xenon, there were other attempts to determine accurately the isotopic composition of xenon trapped in meteorites<sup>3-5</sup>. All of these studies<sup>1-5</sup> concluded that the xenon trapped in average carbonaceous chondrites (AVCC Xe) may be a mixture of fractionated atmospheric-type xenon plus a fission component. However, the concept of AVCC Xe as primordial, when defined as a mixture of two components, was unclear, and this concept has become increasingly vague as the results of stepwise heating experiments on different carbonaceous chondrites in several laboratories<sup>6-13</sup> convincingly demonstrate that the two components of AVCC xenon are inhomogeneously mixed.

It seemed that accurate measurements on the isotopic composition of xenon from the sun might help elucidate the nature of primordial xenon in our solar system, and a component of xenon from the solar wind has recently been identified in the Pesyanoe meteorite<sup>14</sup> and in lunar soil<sup>15-20</sup>. The general features of this solar wind xenon can be related to atmospheric xenon by isotopic mass fractionation, although some of the xenon isotopes,  $^{129}\text{Xe}$ ,  $^{134}\text{Xe}$ ,  $^{136}\text{Xe}$  and  $^{124}\text{Xe}$  or  $^{126}\text{Xe}$ , consistently deviate from the general fractionation pattern<sup>19-22</sup>. It has been suggested that deviations of these isotopes from the general fractionation pattern are due to radiogenic  $^{129}\text{Xe}$  in the atmosphere<sup>22</sup>, to

fissiogenic  $^{134}\text{Xe}$  and  $^{136}\text{Xe}$  in lunar fines<sup>20,22</sup>, and to either an excess of  $^{126}\text{Xe}$ , or a deficiency of  $^{124}\text{Xe}$ , in solar xenon due to nuclear reactions<sup>20,23</sup>.

In a recent report<sup>24</sup> we noted that the xenon released from carbonaceous chondrites at  $\approx 600^\circ\text{--}1000^\circ\text{C}$  contains an excess of the proton-rich isotopes,  $^{124}\text{--}^{128}\text{Xe}$ , in addition to an excess of the neutron-rich isotopes,  $^{131}\text{--}^{136}\text{Xe}$ . It was suggested that trapped meteoritic xenon consists of two isotopically distinct components, and the component which is enriched in both the light and the heavy isotopes relative to  $^{130}\text{Xe}$  was referred to as X. In this report, the available data on xenon in carbonaceous chondrites, in lunar fines, and in the earth's atmosphere are examined in the framework of this component X in seeking a better understanding of the nature of trapped meteoritic xenon.

#### General Isotopic Anomaly Patterns of Xenon

Before discussing the isotopic anomalies of xenon which are observed in different temperature fractions when this gas is released from meteorites by stepwise heating experiments, it is instructive to compare the isotopic composition of this element in the three major reservoirs of xenon which have been studied; xenon in the earth's atmosphere, in meteorites, and in the solar wind. The isotopic composition for xenon in these sources is presented in Table 1. The isotope,  $^{129}\text{Xe}$ , is excluded from consideration here due to its production<sup>1</sup> in the early solar system by the decay of primordial  $^{129}\text{I}$ .

In Table 1, the nonradiogenic and nonspallogenic isotopes of xenon in lunar fines are used to represent the composition of solar wind xenon. The results from various laboratories are shown in Table 1 because estimates on the composition of xenon implanted in lunar fines differ significantly from each other and there is no unanimity of opinion on the actual isotopic composition of xenon in the solar wind. However, these various estimates of



solar xenon show a systematic relationship which suggests that they may be related by mass-dependent fractionation<sup>20</sup>.

Two estimates for the composition of xenon in meteorites are represented in Table 1 by AVCC and WAVCC. The values shown for AVCC are an average of the estimate of trapped meteoritic xenon based on 7 meteorites analyzed by Marti<sup>4</sup> and 3 carbonaceous chondrites analyzed by Eugster *et al.*<sup>5</sup>. The values shown for WAVCC represent a weighted average for approximately 75 analyses in different laboratories on a total of 35 carbonaceous and gas-rich meteorites, as listed in Table 5 of Mazor *et al.*<sup>26</sup>. These results were weighted according to the xenon content of the meteorites. The abundances of the two light xenon isotopes, <sup>124</sup>Xe and <sup>126</sup>Xe, are not shown in WAVCC due to the absence of data on these isotopes for many of the analyses<sup>26</sup>. The lower abundance of the heavy xenon isotopes in WAVCC than in AVCC indicates that the relative abundance of the heavy isotopes is lower in xenon-rich meteorites than in average carbonaceous chondrites. For atmospheric xenon the results of the widely accepted analysis by Nier<sup>25</sup> and a recent analysis by Podosek *et al.*<sup>18</sup> are shown in Table 1.

To compare the composition of xenon in these three reservoirs, we employ the method of Canalas *et al.*<sup>27</sup>, where the abundance of each isotope of mass number,  $i$ , is compared to its atmospheric abundance by the factor,  $F^i$ , defined as follows:

$$F^i = ({}^i\text{Xe}/{}^{130}\text{Xe})_{\text{sample}} / ({}^i\text{Xe}/{}^{130}\text{Xe})_{\text{air}} \quad (1)$$

The values of  $F^i$  for solar and AVCC xenon are plotted against mass number,  $i$ , in Fig. 1. For this comparison we have used Nier's value for atmospheric xenon<sup>25</sup> and Kaiser's value for solar xenon<sup>19</sup>. The latter was selected because it represents an extreme of the estimates for solar xenon and it has been earlier depicted as solar xenon with the lowest fission component.

Several investigators<sup>14-17</sup> have suggested that solar xenon and AVCC xenon are identical, except for the presence of excess  $^{132-136}\text{Xe}$  in the latter. It had been noted earlier that atmospheric xenon and AVCC xenon could be related by mass dependent fractionation<sup>1-5</sup>, except for the presence of excess  $^{132-136}\text{Xe}$  in the latter. Thus, the view of solar and AVCC xenon expressed by these investigators<sup>14-17</sup> suggests the possibility that atmospheric and solar xenon are directly related by fractionation. Kaiser<sup>19</sup> has recently interpreted his results on xenon in Luna 16 as evidence that atmospheric xenon is, in fact, fractionated solar xenon. However, as can be seen in Fig. 1, even for this extreme estimate of solar xenon given by Kaiser<sup>19</sup>, there is steeper fractionation across the light isotopes,  $^{124-131}\text{Xe}$ , than across the heavy isotopes,  $^{132-136}\text{Xe}$ . The break in the fractionation patterns for both AVCC and solar xenon at  $^{132}\text{Xe}$  clearly demonstrates that neither of these two reservoirs of extraterrestrial xenon can be related to atmospheric xenon by simple mass dependent fractionation.

#### Component X and Trapped Meteoritic Xenon

In 1964 Reynolds and Turner<sup>6</sup> reported the presence of excess heavy xenon isotopes in the gas released from the Renazzo carbonaceous chondrite at 700°-1000°C. Stepwise heating experiments on other carbonaceous chondrites in several laboratories<sup>7-13</sup> confirmed this anomaly. Last year<sup>24</sup> we reported that the results of stepwise heating experiments showed an enrichment of  $^{124}\text{Xe}$  in the same temperature fractions which contained excess  $^{136}\text{Xe}$ . The correlation observed between light and heavy isotopes could not be explained by any of the previously proposed mechanisms, and we concluded that the observed pattern of anomaly was due to the release of an isotopically distinct component of xenon, referred to as component X, which had been incorporated into the meteorites.

Table 1. Isotopic Composition of Solar, Meteoritic and Atmospheric Xenon

Sample	$^{124}\text{Xe}$	$^{126}\text{Xe}$	$^{128}\text{Xe}$	$^{130}\text{Xe}$	$^{131}\text{Xe}$	$^{132}\text{Xe}$	$^{134}\text{Xe}$	$^{136}\text{Xe}$	References
<u>Solar:</u>									
Luna - 16	2.99	2.78	51.0	100	494.7	600.6	220.1	176.0	Kaiser <sup>19</sup>
Lunar Fines - 10084.59	3.02	2.93	51.5	100	498.0	606.0	220.0	179.0	Hohenberg <u>et al.</u> <sup>15</sup>
Lunar Fines - 10084.48	2.94	2.55	50.8	100	496.0	608.0	224.2	182.6	Pepin <u>et al.</u> <sup>17</sup>
Lunar Fines - SUCOR	2.89	2.63	50.9	100	499.0	606.6	225.2	182.7	Podosek <u>et al.</u> <sup>18</sup>
Lunar Fines - 10084.47	2.99	2.67	50.45	100	495.5	607.0	225.8	184.2	Eberhardt <u>et al.</u> <sup>16</sup>
Lunar Fines - 15601.64	2.73	2.61	50.6	100	501.2	613.5	229.4	187.1	Srinivasan <u>et al.</u> <sup>20</sup>
<u>Meteoritic:</u>									
AVCC	2.828	2.533	50.38	100	506.7	620.9	236.9	199.0	Marti <sup>4</sup> and Eugster <u>et al.</u> <sup>5</sup>
WAVCC	--	--	50.82	100	506.3	617.9	235.5	197.2	This Work
<u>Atmospheric:</u>									
Air	2.353	2.206	47.03	100	519.1	659.1	255.9	217.4	Nier <sup>25</sup>
Air	2.334	2.175	47.07	100	522.4	661.4	256.7	218.2	Podosek <u>et al.</u> <sup>18</sup>

Table 2. Anomalous Xenon Released in Stepwise Heating of the Carbonaceous Chondrites

Sample	$^{124}\text{Xe}$	$^{126}\text{Xe}$	$^{128}\text{Xe}$	$^{130}\text{Xe}$	$^{131}\text{Xe}$	$^{132}\text{Xe}$	$^{134}\text{Xe}$	$^{136}\text{Xe}$	References
Renazzo 800°	2.93±0.05	2.57±0.03	50.6±0.4	100	505.8±2.4	617.2±1.9	243.3±2.2	210.4±1.5	Reynolds & Turner <sup>6</sup>
Leoville 1010°	3.07±0.09	2.66±0.12	51.4±0.5	100	511.3±2.4	625.0±3.9	250.0±2.4	220.6±2.3	Manuel <u>et al.</u> <sup>11</sup>
Mokoia 700°	3.43±0.05	3.02±0.06	51.9±0.4	100	512.0±2.5	625.0±2.4	269.7±1.3	244.8±1.2	Phinney <sup>13</sup>
Mokoia (matrix) 600°	3.15±0.09	2.72±0.09	52.7±0.7	100	519.9±6.8	633.3±7.6	276.1±3.6	248.9±3.6	Rowe <sup>9</sup>
Mokoia 750°	3.39±0.06	2.90±0.05	52.0±0.8	100	514.1±5.6	624.5±5.6	278.6±3.2	261.1±2.0	Manuel <u>et al.</u> <sup>12</sup>
Mokoia 800°	3.44±0.04	2.83±0.04	52.2±0.2	100	514.8±1.8	628.9±1.9	280.8±1.0	263.6±1.1	Phinney <sup>13</sup>
Allende 700°	3.37±0.10	2.93±0.10	50.7±0.5	100	516.3±3.2	633.7±3.5	289.2±2.7	273.6±3.0	Phinney <sup>13</sup>
Allende 1000°	3.39±0.14	2.77±0.09	54.5±0.9	100	513.0±6.1	624.9±8.6	291.2±3.9	282.5±2.4	Manuel <u>et al.</u> <sup>12</sup>
Allende 600°	3.42±0.09	3.02±0.12	52.1±0.9	100	522.7±5.6	643.1±6.2	300.8±5.0	290.0±3.4	Phinney <sup>13</sup>
Allende 800°	3.62±0.13	2.84±0.09	52.1±1.5	100	518.6±11.9	641.0±14.3	305.1±7.0	299.4±5.4	Manuel <u>et al.</u> <sup>12</sup>

Since our initial report was published, the results of analyses by Phinney<sup>13</sup> on xenon released by stepwise heating of carbonaceous chondrites have been brought to our attention. These results, for the temperature fractions showing the largest enrichment of  $^{136}\text{Xe}$ , are listed in Table 2 together with the results of stepwise heating experiments discussed in our earlier report<sup>24</sup>.

The correlation between the enrichment of the light isotopes,  $^{124}\text{Xe}$  and  $^{126}\text{Xe}$ , and the enrichment of the heaviest isotope,  $^{136}\text{Xe}$ , in the xenon released by stepwise heating can be seen in Fig. 2, where  $^{124}\text{Xe}/^{130}\text{Xe}$  and  $^{126}\text{Xe}/^{130}\text{Xe}$  are plotted against  $^{136}\text{Xe}/^{130}\text{Xe}$ . Estimates of solar, atmospheric, and meteoritic xenon from Table 1 are shown for comparison. As can be seen from Fig. 2, the recent data by Phinney<sup>13</sup>, which is represented by the darkened points, agrees with the correlation reported earlier<sup>24</sup>. It should be noted that the abundances of  $^{124}\text{Xe}$  and  $^{126}\text{Xe}$  have not been corrected for cosmic-ray induced spallation reactions, although we suspect that the scatter of data points from the correlation lines may be partially due to the presence of a cosmogenic component.

The dashed line in Fig. 2 shows the relationship expected from diffusive fractionation<sup>28</sup>, and the failure of solar xenon to lie on the fractionation line passing through atmospheric xenon is understood in terms of the relationship shown in Fig. 1. The solid line connecting atmospheric and solar xenon defines the isotopic composition of xenon which would be produced by mixing components from these two reservoirs of xenon. This "mixing" line approximately parallels the fractionation relationship, and it appears that subtraction of component X from the xenon released at each extraction temperature might leave a common trapped meteoritic xenon component. If AVCC Xe properly represents an average

of the total xenon in meteorites, then the composition of trapped meteoritic xenon, when corrected for the presence of component X, is expected to lie to the left of AVCC Xe on the correlation lines shown in Fig. 2. Since the other two known reservoirs of xenon are solar and atmospheric, a suitable choice for trapped meteoritic xenon can be characterized by the intersections of the correlation lines with the "mixing" line or the fractionation line. The darkened column labeled trapped meteoritic in Fig. 2 represents these intersections. Until the isotopic composition of solar xenon is sufficiently well established to clearly indicate the relationship between solar and atmospheric xenon, a decision on which intersection represents trapped meteoritic xenon would be arbitrary. Since these three reservoirs of xenon are probably genetically related, though not necessarily by a diffusive fractionation relationship, we will employ a simple linear relationship between atmospheric and solar xenon to see if meteoritic xenon can be resolved into a mixture of component X and a self-consistent composition for trapped meteoritic xenon.

The observed correlations between the other non-radiogenic xenon isotopes released in the stepwise heating of carbonaceous chondrites are shown in Fig. 3, together with the "mixing" line between solar and atmospheric xenon. All of the xenon isotope data from Table 1 and Table 2 for  $i = 128-136$  are shown in Fig. 3, except for the ratio,  $^{128}\text{Xe}/^{130}\text{Xe}$ , in the xenon released from Allende at  $1000^\circ\text{C}$ . The unusually high value for this ratio, indicated by the arrow in Fig. 3, is attributed to neutron-capture reactions on  $^{127}\text{I}$  in this meteorite. Fireman et al.<sup>29</sup> and Manuel et al.<sup>12</sup> have reported an excess of certain noble gas isotopes in Allende due neutron-capture reactions on halogens.

From Fig. 3 it appears that the points of intersection of the correlation lines and the "mixing" lines define a self-consistent composition for trapped

meteoritic xenon, and this composition is also consistent with the trapped xenon shown in Fig. 2. The isotopic composition of trapped meteoritic xenon defined by these intersections is;  $^{124}\text{Xe} : ^{126}\text{Xe} : ^{128}\text{Xe} : ^{130}\text{Xe} : ^{131}\text{Xe} : ^{132}\text{Xe} : ^{134}\text{Xe} : ^{136}\text{Xe} = 0.0276 : 0.0248 : 0.501 : 1.00 : 5.04 : 6.19 : 2.31 : 1.90$ . The scatter of data points about the correlation lines in Fig. 3 may be due to variations in the composition of trapped meteoritic xenon. If the trapped meteoritic xenon lies along the "mixing" line, as would be expected if meteoritic xenon has been acted on by the same process responsible for the relationship between atmospheric and solar xenon, then this might account for the smallest scatter of data points about the correlation line for  $i = 134$ . For this isotope, the slope of the "mixing" line and the slope of the correlation line are most nearly equal.

It should be noted from Fig. 3 that, relative to  $^{130}\text{Xe}$ , all of the other xenon isotopes are enriched in component X when compared to their abundance in trapped meteoritic xenon. We have discussed possible origins for component X earlier and have no additional suggestions to make for its source. However, a few comments on the relationship between AVCC Xenon, trapped meteoritic xenon, and component X seem appropriate.

#### AVCC Xenon

Mazor et al.<sup>26</sup> have shown the results of approximately 75 analyses for meteoritic xenon from several laboratories. These results on the total xenon do not include data on  $^{124}\text{Xe}$  and  $^{126}\text{Xe}$  for most of the meteorites, and no data on  $^{130}\text{Xe}$  is given for 4 analyses. Thus, it is not possible to determine if the total xenon in these meteorites shows a correlation between an enrichment of the light isotopes and an enrichment of the heavy isotopes, as shown in Fig. 2 for stepwise heating experiments.

However, even in total melt experiments, we find variations in the relative abundances of the heavy xenon isotopes in the manner observed for stepwise heating experiments. This correlation of excess  $^{136}\text{Xe}$  with excess  $^{134}\text{Xe}$  in about 70 total melt analyses of the xenon in 35 different meteorites is shown in Fig. 4. The dashed lines indicate the area of uncertainty for the correlation which would result if points on the solid correlation line had an uncertainty of  $\pm 1\%$ . This degree of uncertainty is typical of the xenon isotope measurements, and this is shown for a symbolic data point in the lower right of Fig. 2. The isotopic compositions given in Table 1 for AVCC and WAVCC are shown on Fig. 4. The fact that these lie near the middle of the graph, which is heavily populated with datapoints representing total meteoritic xenon, demonstrates that AVCC and WAVCC well represent the average isotopic composition of the total meteoritic xenon.

Nevertheless, the correlation of excess  $^{134}\text{Xe}$  with excess  $^{136}\text{Xe}$  in the manner observed for xenon released by stepwise heating experiments clearly shows that variations in the isotopic composition of the total meteoritic xenon are probably due to variations in the relative amounts of component X. The range of values for the relative amounts of component X indicated by Fig. 4 does not agree with an earlier report<sup>30</sup> that excess  $^{136}\text{Xe}$  displays a  $45^\circ$  correlation with the xenon content of meteorites. In view of this discrepancy, caution should be exercised regarding conclusions based on the earlier estimate<sup>30</sup> of excess  $^{136}\text{Xe}$  in meteorites. These would include estimates of the half-life<sup>30</sup>, the atomic number<sup>30,31</sup>, the heat of vaporization<sup>31</sup>, and the boiling point<sup>31</sup> of some hypothetical element which might have produced the excess  $^{136}\text{Xe}$  by fission.

From the range of  $^{136}\text{Xe}/^{130}\text{Xe}$  values shown in Fig. 3 and Fig. 4 we conclude that for total melt experiments the  $^{136}\text{Xe}/^{130}\text{Xe}$  ratio may differ from the  $^{136}\text{Xe}/^{130}\text{Xe}$  ratio in trapped meteoritic xenon by  $\approx 0-11\%$  due to variations in



the relative amounts of component X, whereas stepwise heating experiments partially separate these two components of meteoritic xenon to yield  $^{136}\text{Xe}/^{130}\text{Xe}$  ratios that are  $\approx 0-58\%$  higher than the  $^{136}\text{Xe}/^{130}\text{Xe}$  ratio in trapped meteoritic xenon. AVCC xenon represents the isotopic composition for the average observed mixture of trapped meteoritic xenon and component X.

This investigation was supported by NASA.

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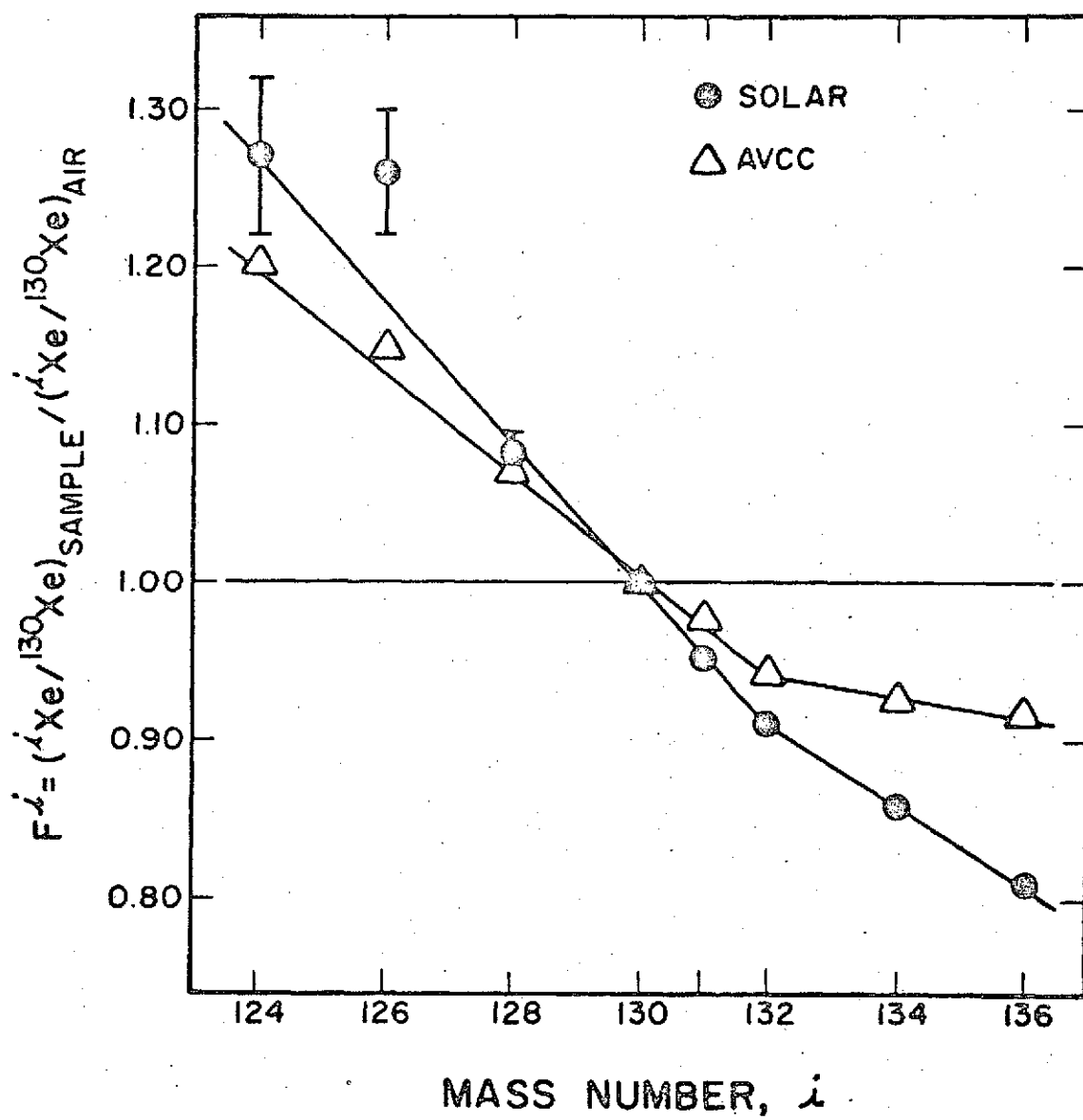
## Figure Captions

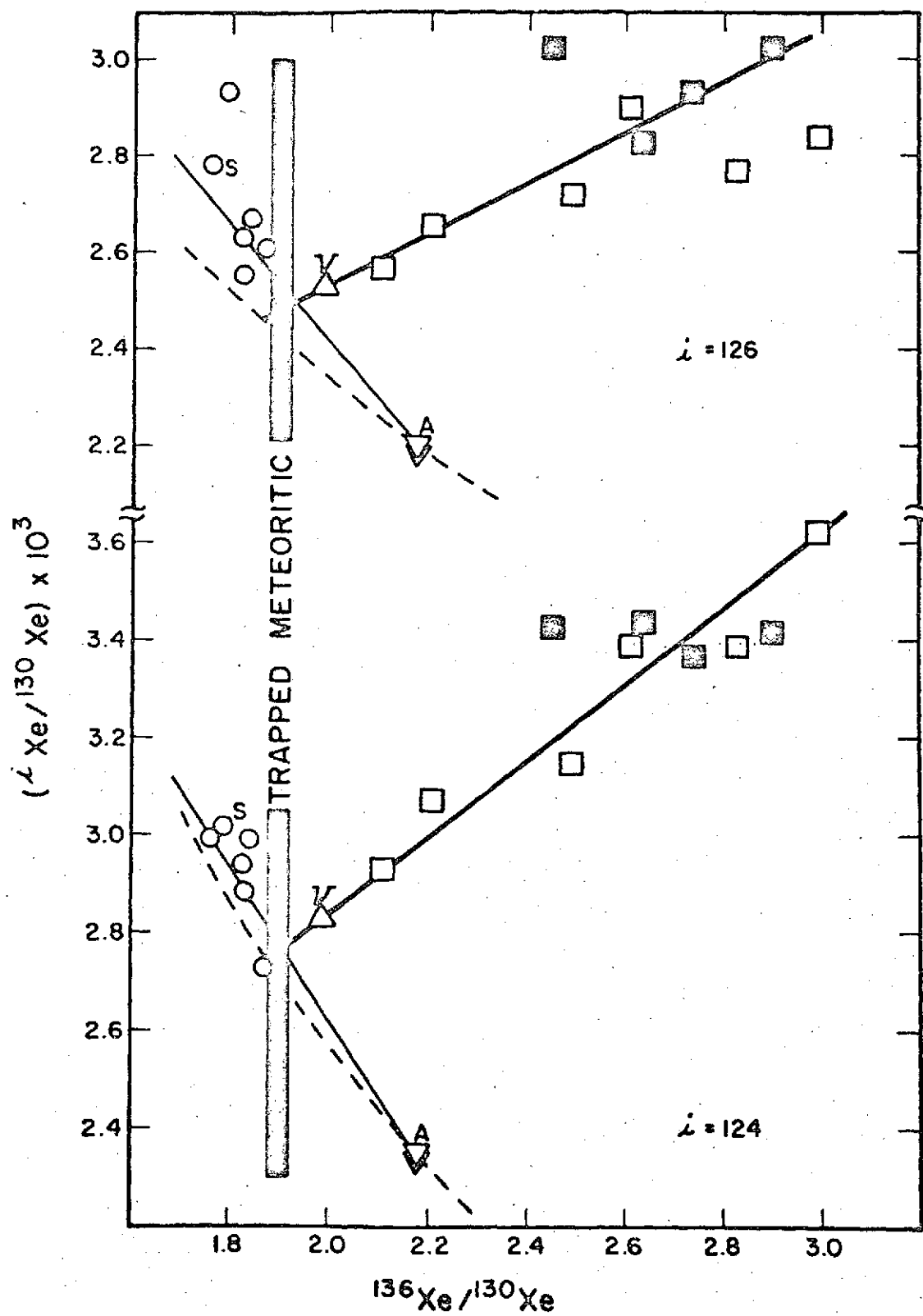
Fig. 1. A comparison of AVCC xenon<sup>4,5</sup> and solar xenon<sup>19</sup> with xenon in the earth's atmosphere<sup>25</sup>. The break in the lines at <sup>132</sup>Xe indicates that neither of these two forms of extraterrestrial xenon can be related to atmospheric xenon by mass dependent fractionation.

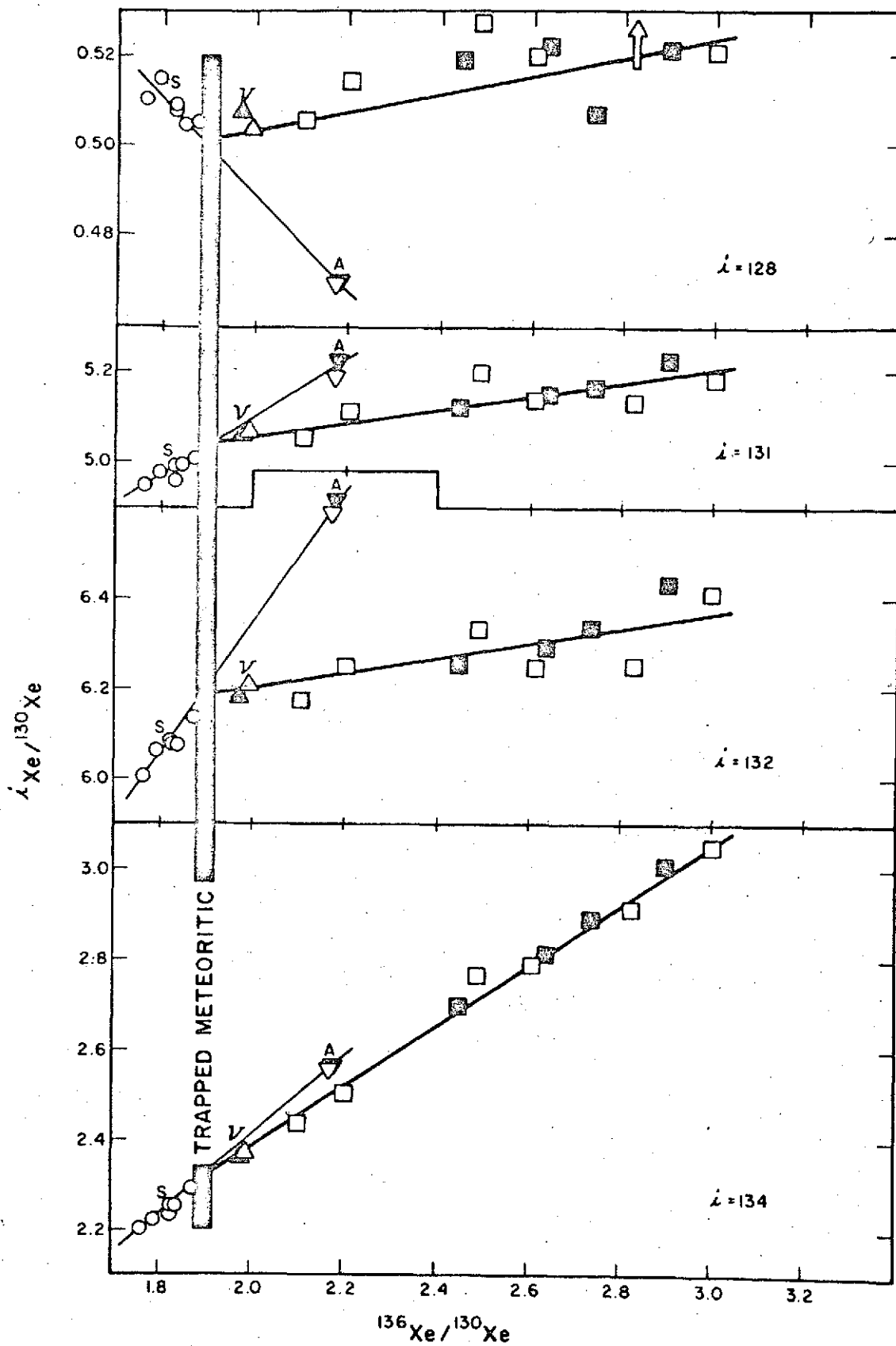
Fig. 2. The correlations of excess <sup>124</sup>Xe and excess <sup>126</sup>Xe with excess <sup>136</sup>Xe in the xenon released by stepwise heating of carbonaceous chondrites. The correlations between these xenon isotope ratios are shown by the heavy lines. Recent stepwise heating data by Phinney<sup>13</sup> are shown as dark squares. The symbols S, V, A are shown above solar, AVCC, and atmospheric xenon, respectively. The dashed line passing through atmospheric xenon shows the isotopic compositions expected from diffusive fractionation, and the solid line passing through atmospheric and solar xenon show the isotopic compositions expected from mixing of these two types of xenon.

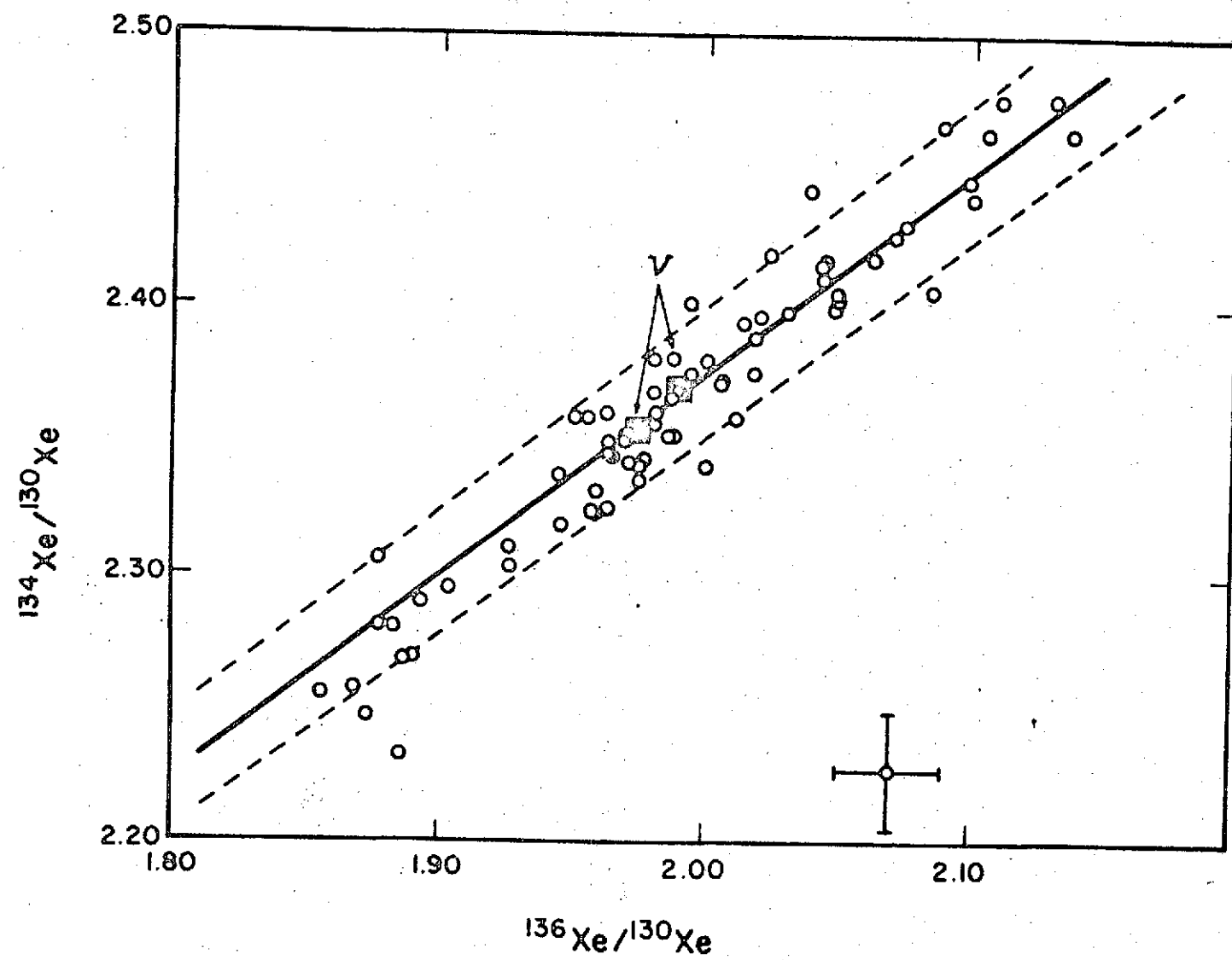
Fig. 3. The correlation of other xenon isotopes with excess <sup>136</sup>Xe in the xenon released by stepwise heating of carbonaceous chondrites. The lines and symbols are as defined for Fig. 2 except that both AVCC and WAVCC are shown below the symbol V.

Fig. 4. The correlation of excess <sup>134</sup>Xe with excess <sup>136</sup>Xe in the total xenon of meteorites. Data are from a compilation by Mazor et al.<sup>26</sup>. The darkened squares, labeled, V, show the isotopic composition of AVCC and WAVCC xenon. Typical error limits of  $\pm 1\%$  are shown in the lower right corner.











## APPENDIX - II

(Paper to be Submitted)

Xenon Anomaly: The Possible Role of Mass Fractionation

D. D. Sabu

Department of Chemistry

Grambling College, Grambling, LA 71245

ABSTRACT. Shale on heating for 95 hours at 200°C shows a similar mass fractionation pattern of xenon isotopes as that exhibited by solar wind relative to atmosphere. This suggests that atmospheric xenon may be a mass fractionated solar wind xenon. A mechanism in terms of early incorporation of solar wind xenon in the primordial matter is proposed to explain the abundance anomaly of Krypton and practically negligible mass fractionation of Kr and Ar in comparison to Xe and Ne isotopes in the earth's atmosphere.

It is well established that earth, moon, and meteorites have lost much of their original abundance of noble gases. What is not established is has any of this loss been accompanied by a mass fractionation process resulting in isotopic anomaly of noble gases. Kuroda and Manuel (1970) believe that the mass fractionation process is responsible for isotopic anomaly of trapped neon and xenon in meteorites. On the other hand the approach taken by Chicago and Minnesota groups is that the isotopic compositions of trapped noble gases particularly He and Ne are mixtures of several component whose origins are not well understood at present (Pepin, 1967; Black and Pepin 1969; Black, 1972; Mazor et al., 1970). There exists a strong possibility that early losses of noble gases from planetary bodies might accompany mass fractionation (Suess, 1949), however attempts to explain the isotopic anomaly of noble gases in terms of mass fractionation process has proved to be a needle in a bundle of hay. The purpose of this note is to bring to attention the results of an experiment done in 1967 by Canalas which further points out the mysterious role the mass fractionation process may have played in the early history of solar system.

Shale is one of the richest source of xenon on earth (Canalas et al., 1968). Its xenon is atmospheric in isotopic composition (Canalas et al., 1968). Canalas (1967) heated a sample of shale for 95 hours at 200°C and analyzed the xenon released at 400°C after heating for one hour. The results of his analysis along with solar wind xenon estimate from Luna-16 (Kaiser 1972) and atmospheric xenon (Nier, 1950) are presented in Table-1.

Using the method of Canalas et al., (1968) the fractionation factor  $F^m$  vs.  $M$  are plotted in Fig. 1 for Shale-400°C fraction and solar wind xenon. The

fractionation factor  $F^m$  is defined as follows:

$$F^m = \frac{(Xe^m / Xe^{130})_{\text{sample}}}{(Xe^m / Xe^{130})_{\text{atmosphere}}}$$

In Fig. 1 Shale-400°C fraction and solar wind xenon exhibit a mass fractionation pattern which is non-linear but similar. The difference being that the two are fractionated in opposite direction with solar wind xenon showing relatively more severe fractionation. The reasons for this are understandable. Shale represents a mass fractionated atmospheric xenon while the atmospheric xenon is most likely a mass fractionated solar wind xenon. The significant fact here is that the pattern of mass fractionation is similar, showing a break in linearity at mass number-132. Assuming there occurs some complex mechanism of mass fractionation by which the observed pattern of Fig. 1 can be explained, then the unsolved puzzle of xenon isotopes anomaly in moon, meteorites and earth can be put in proper perspective. Recently Sabu et al., (1973) showed that the trapped xenon in carbonaceous chondrites is different from average carbonaceous chondrite (AVCC) xenon. AVCC xenon is a mixture of trapped xenon and component X xenon (Manuel et al., 1972). Sabu et al., (1973) showed that the trapped xenon in carbonaceous chondrites is related to solar wind xenon by the same process which relates atmospheric xenon to solar wind xenon. Thus mass fractionation seems to be a prominent process by which the isotopic composition of trapped meteoritic, solar wind and earth's atmospheric xenon can be interrelated.

An apparent difficulty that arises from any of the mass fractionation mechanisms

is the similarity of isotopic composition of atmospheric Ar and Kr to that of solar wind (Eberhardt et al., 1970) and trapped meteoritic Ar and Kr (Mazor et al., 1970). Following model is proposed to explain the negligible mass fractionation of earth's Kr and Ar relative to its xenon:

Assuming the losses of primordial noble gases from the primitive planetary material have occurred to a larger extent than earlier believed (Suess, 1949). About 25% of xenon found on earth remained in the primordial matter which subsequently formed the planet earth. Further assuming that the losses of noble gases from the primordial matter were accompanied at least in part by a severe mass fractionation. As a result of this loss, Xe, Kr, Ar, Ne and He became severely mass fractionated. However the concentration of other noble gases except xenon in the primordial matter will become negligible. At this stage if the primordial matter is still in the form of submicron size grains then it will effectively absorb solar wind noble gases. In reality the loss of noble gases and implantation of solar wind can occur simultaneously. Under such conditions there will occur a stage of equilibrium when the solar wind influx of noble gases become equal to outflux of these gases. Earliest to reach this equilibrium stage would of course be He then Ne etc. In the same sequential order these noble gases will assume the solar isotopic composition. Meteorites such as Pesyanoe contain solar type noble gases (Marti, 1969; Eberhardt et al., 1965; Sabu 1973) and other evidences (Lal and Rajan, 1969; Pellas et al., 1969) strongly suggest that primitive matter was exposed to solar wind in its early history. Calculations regarding duration of this exposure are highly model dependent and beyond the scope of the present paper. Assuming the exposure was long enough so that xenon on earth is

approximately a mixture of 75% solar wind and 25% severely mass fractionated xenon. The calculation of the ratio of severely fractionated Kr to solar Kr depends upon the Kr/Xe ratio at the termination of loss of noble gases. Assuming  $(\text{Kr}^{84} / \text{Xe}^{132})_{\text{fractionated}}$  equals 0.1 then less than 0.5% of earth's krypton is severely fractionated. The evidence that earth's krypton is predominately solar in origin comes from the present abundance of Kr/Xe in its atmosphere.  $\text{Kr}^{84} / \text{Xe}^{132}$  ratio in earth's atmosphere is even higher than relative cosmic abundance as estimated by Cameron (1969). We may try to explain the anomalous abundance of atmospheric krypton relative to its xenon by assuming a slight outgassing of earth's xenon. Assuming at the time of formation of earth the Kr/Xe ratio was similar to chondrites then one arrives at 4% outgassing of earth's xenon for 100% outgassing of its krypton. This represents a high degree of separation between Kr and Xe and even on charcoal such high degree of separation can not be easily obtained. A better explanation for the anomalous abundance of Kr in earth's atmosphere can be offered in terms of partial outgassing of earth along with high Kr/Xe ratio in earth at the time of its formation due to incorporation solar wind as discussed in the model above.

The abundance of the He, Ne, and Ar in earth's atmosphere are not anomalously high. In the course of grain growth and the formation of planetesimals a significant loss of light noble gases is expected. Some of this loss may accompany diffusive fractionation. Most likely mass fractionation of Ne occurred in this manner. Secondary losses of Ar are not likely to occur to any large extent. Consequently argon like Kr exhibits an insignificant pattern of mass fractionation. Helium on earth is fractionated because of the secondary losses and due to gravitational escape.

Acknowledgements - The author is grateful to O. K. Manuel for pointing out the experiment done by Canalas in 1967. This work was supported by National Aeronautics and Space Administration Grant NGR 19-011-012.

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TABLE 1.

Isotopic composition of xenon released from shale at 400° C after 95 hours of heating at 200° C.

Mass Number	Shale - 400° C	Atmosphere	Solar Wind
m	Canalas (1967)	Nier (1950)	Kaiser (1972)
124	0.0225±0.0002	0.02353	0.0299±0.0012
126	0.0212±0.0002	0.02206	0.0278±0.0009
128	0.461±0.002	0.4703	0.5101±0.055
129	6.434±0.031	6.480	6.338±0.065
130	1.000	1.000	1.000
131	5.256±0.022	5.191	4.947±0.030
132	6.732±0.023	6.591	6.006±0.030
134	2.616±0.01	2.559	2.201±0.018
136	2.252±0.01	2.174	1.760±0.020

### Figure Caption

Fig. 1. Fractionation factor  $F^m$  vs. mass of xenon isotopes for shale and solar wind. Both show a break in linear pattern at mass 132.

